Mechanical Disturbances

Mechanical, or unnatural, disturbances were also studied as a potentially important resuspension mechanism. This includes such activities as mowing, well drilling, construction, digging, or soil removal. Mowing was considered of major interest since it takes place every year and covers the whole area. Mowing involves disturbance of both grass and soil. While research sampling took place in the 903 Field east of the plant security fence, the grass was cut during dry conditions. This was expected to maximize resuspension. At times the mowing tractor would pass right next to an air sampler. A statistically significant increase by a factor of 5 in the total Pu concentration was discerned during such a period in June 1981 (RF3464, p4). But no change was found during a similar period in 1983 at the same location (LA86, p90). Again, high variability in Pu concentrations make trends difficult to establish unless many samples are taken. In June and July of 1987, wells were drilled in the 903 Field and the nearby surveillance air samplers showed an increase by a factor of about 3 in Pu radioactivity, but such increases are often seen during the dry summer months regardless of soil disturbances. Mechanical disturbances are short-term events.

Consideration was also given to resuspension of dust containing Pu from two unpaved roads intersecting the 903 Field along the security fence. It is evident from the color and quantity of the dust collected by samplers located along the roads that much of it is resuspended by traffic. A 1973 study (MI73) showed that road dust radioactivity averaged 68 pCi/gm and remedial action was initiated (e.g., oiling, grading, etc). In 1980 another road dust evaluation was carried out. As a first step the Pu radioactivity of the road surface for one of the roads was determined by conventional soil sampling methods (RF3689, p18). The Pu radioactivity of this road surface was surprisingly low, 4.6 pCi/g of Pu-239, when compared to adjacent soils of 790 pCi/g of Pu-239 that were monitored a distance of one foot from the road. However, since 1973 the road was often graded and ballast added. It had been expected that the adjacent soil would provide a source of Pu for easy resuspension by traffic on the dirt road. However, the adjacent soil was not disturbed by traffic on the road.

Also in the 1980 evaluation a truck was driven along this road to complete the comparison with the 1973 test, and to sample a larger area than that covered by a few road soil samples. The dust generated behind a rear wheel was sampled with a high-volume air sampler (hivol) similar to that used for the dispersion studies described later. The device gives a

<3-μm and >3-μm cut. The road dust was very low in radioactivity, measuring 6.0 pCi/g of Pu-239 compared to 1000 and 2000 pCi/g of Pu-239 for two adjacent soil areas. It was concluded that the roads are no longer a significant problem, especially because of the low volume of traffic. Of considerable interest was the incidental new information that no measurable amounts of respirable (<3-μm AED) particles are generated (RF3287, p7). This was also observed during a recent dam construction project at RFP. Heavy earth moving machinery created no additional respirable dust (RF3115, p6).

Such observations indicate that considerable application of force is necessary to create <3-μm AED particles. It has been noticed that the particles <3-μm AED were mostly black combustion particles generated by vaporization-condensation (e.g., combustion) by vehicles, furnaces and other industrial activity. This is in line with the wind tunnel results, that soil resuspended from the field is very low in respirable particles, those less than 3 μm in size (RF3689, p35) and many of these particles found at RFP are a result of Denver pollution (RF3990, p31).

DISPERSION OF Pu PARTICLES FROM RFP

Combining the above processes into a coherent, predictive Pu transport model is obviously beset with problems, especially with the resuspension from the 903 Field being relatively low and of varying nature (wind, rain splash and release from grass). Procedures from previous studies can be used to derive conventional resuspension parameters which are commonly used to provide a rough estimate of the emission of soil particles containing contaminants (HA80a, p210). Such parameters are given in the section titled: "Resuspension Factors for Pu Release from the 903 Field" in this report. However, site-specific experiments were necessary for realistic estimates of Pu dispersion from RFP which would directly measure the emission and transport of Pu particles. These experiments included measurements of the vertical distribution of Pu particles in the air that passes over the 903 Field.

Two distinct steps were involved in these experiments. First, an attempt was made to measure the Pu particle flux from the 903 Field at the source. A number of research air samplers were deployed at selected points in the field to determine the total resuspension of Pu. Second, a vertical array of samplers was installed, at some distance from the 903 Field, to measure the Pu particle concentration in the air that passed over the field.

Determination of Pu Flux Characteristics for Source Area

For the first step, four research hivols (~1 m^3/min) were installed toward the perimeter areas of the 903 Field and an ultrahigh-volume air sampler (ultravol - ~7m^3/min) was installed near the center of the field to determine how much Pu is being released to the environment. The ultravol (Figure 10), operating at 7.4 m^3/min. and providing a <10-μm and >10-μm fraction, was changed weekly or more often to provide high resolution Pu concentration data (RF3197, p6). This was used for special events such as wind storms, periods of snow or rain and fallout from atmospheric nuclear weapons tests (RF3464, p5). The research hivol samplers provided <3-μm AED and >3-μm AED fractions. These corresponded to a respirable and an inhalable-plus coarse particle size cut, respectively. The >3-μm AED fraction was collected by combining the particles collected by a cyclone pre-separator with a nominal 5-μm cut, followed by two impactor stages to provide a sharp 3-μm cut. The cyclone sampler inlet was designed to turn with the wind so that the intake always pointed into the wind. The efficiency of the cyclone was evaluated and one

observation was that the inlet efficiency was not sensitive to wind speed, that is, the particle concentration and size distribution was unaffected by wind speed (RF3464, p34).

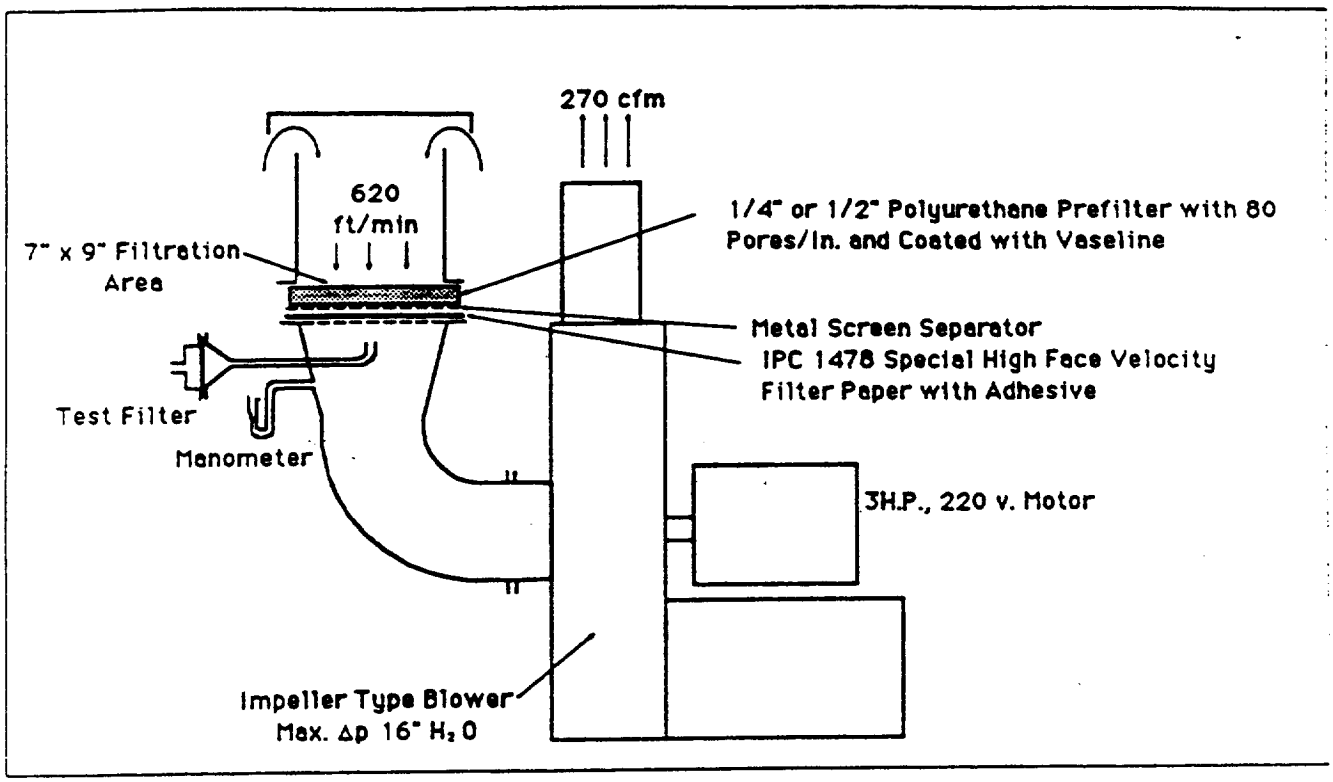

Figure 10. Ultrahigh-volume Air Sampler

The results of approximately two years of sampling (RF3464, p4; RF3650, p1) showed that monthly Pu concentrations in air that passed over the 903 Field varied by a factor of 10 to 100 at a given sampler location over a two year period. No correlation between wind speed and Pu concentration was found. Nor was there any correlation between the Pu concentrations found at the four sampler locations when plotted against time*. This observation even held for another experiment with three samplers at the same location (RF4036, p17, 23). By contrast, the corresponding dust concentrations for these tests varied by a factor of two during the same period and the concentration values at each sampling station showed identical trends with time.

* The same was true for the three RFP surveillance samplers spaced across the pad field along the security fence, i.e., the concentration data between the three samplers did not correlate in time (RF3914, p4). That is, if one sampler had a high concentration the others showed no corresponding increase, etc. The data examined were for a 9-year period.

Naturally, it is desirable know why the Pu concentrations in air at the source area varied so randomly. Especially in view of the fact that the concentration of another radioactive trace material, Be-7, correlated very well between all the samplers versus time (RF3287, p31). Be-7 is of stratospheric origin and consists of near atomic-size particles, which become attached to many dust particles in the troposphere. It is important to note that Be-7 is determined at the same time as the Am-241 values by gamma spectroscopy. The Am-241 gamma spectroscopy values fluctuated widely but correlated with the Pu values determined by alpha spectroscopy, confirming the accuracy and precision of the Pu analytical procedures (RF3650, p5-6). The next step was to examine data on Pu particle size on the presumption that the Pu particles may be relatively large and therefore few in number.

Unfortunately, data on the size of airborne Pu particles at the 903 Field are lacking. Alpha track analysis of an 8" x 10" filter proved to be quite tedious. Only a few tracks per square centimeter were present and most of these were single tracks, indicative of uranium. It was estimated from the multiple tracks that the Pu particles ranged from 0.08 to 0.3 μm (RF3115, p17), but the counting statistics were poor at a × 100 microscopic magnification (i.e., alpha tracks were seldom seen). Wind tunnel resuspension of surface soil particles in the 903 Field (i.e., Pu sources area) revealed that the most common size was 0.06 to 1 μm (WA82, p23). This work was much more definitive because many alpha tracks were present in a microscope counting field.

Finally, Pu particle size data from the soil itself are of interest. Whicker (WH79) reported that most particles were 1 to 2 μm in size. Langer (RF3990, p43) reported Pu particles up to 10 μm in size. An interesting scanning electon microscopy study (ME90, p48-55) by EG&G Idaho of Pu-contaminated soil from Rocky Flats led to the identification of soil particles as large as 30 μm that contained Pu. This confirmed the idea that Pu is carried by host particles. This work also indicated that these large particles are fragile agglomerates.

From the above data it is concluded that the airborne Pu particles sampled were a few discrete, relatively large particles in a huge volume of air. This is very unlike sampling a continuum of a trace gas or atomic clusters of Be-7 attached to many dust particles. To illustrate this point, for a relatively high Pu concentration of 1.4 fCi/m^3 in the 903 Field, 125 Pu particles 1 μm in size would account for the activity found in 40,000 cubic meters of air sampled during a typical run; or, a single 5-μm particle could account for all the activity. Such poor sampling statistics make it impossible to establish short term

concentration trends. That is, at best one can statistically compare only yearly or longer averages from each 903 Field sampling station for meaningful trends. For the community samplers, miles away from the 903 Field, it is impossible to establish Pu trends that relate to RFP activities.

The data cited also showed that most of the Pu activity is on particles >3 μm. To better define the Pu activity vs. particle size, large dust samples (gram-sized, as found during dust storms) were subjected to wet sieving of the >3-μm AED particles into 44 and 74 μm sieve cuts. Freon was used for the suspension medium to prevent de-agglomeration (HA80a, p219). This sieving showed that the Pu distribution is roughly proportional to dust particle mass (RF4036, p22). More accurately, the specific radioactivity of the Pu in the samples was three times higher for the >3 μm fraction than for the respirable fraction. But the specific radioactivity for the respirable fraction was small and not as accurately determined.

During normal wind conditions another test series gave the following results. The concentration of the respirable Pu fraction, 0.02 fCi/m^3 of Pu-239, was nearly at the fallout level of 0.018 fCi/m^3 of Pu-239 for June 1980 to June 1981 (RF3650, p6). The >3-μm AED particle radioactivity was 0.71 fCi/m^3 of Pu-239. There is little emission of <3-μm AED particles from the 903 Field (RF3464, p4). The >3-μm AED fraction carried 97 percent of the radioactivity for the above period.

As an aside, in the spring, large amounts of pine pollen were blown from the foothills over the plant and were collected by the air samplers. Pollen is designed by nature to float long distances due to small air sacs. Scanning electron microscope photos of the coarse particle dust fractions showed the presence of pollen (RF3990, p28) which causes a yellow/green discoloration of dust collected in early summer. No unusual increase in Pu activity was seen during these periods, as suggested by Nichols (NI74).

Dispersion of Pu by Air Passing Over the 903 Field

The above studies were followed by an air sampling program using a scaffold 10 m in height and located 100 m from the eastern edge of the 903 Field (RF3914, p16). The objective was to follow the transport of Pu from the 903 Field. The scaffold had hivol samplers at a height of 1, 3 and 10 m to sample the air that passed over the 903 Field for Pu particles. The three hivol samplers had EPA-researched size selective inlets (SSI) with a

cut of 15-μm AED particle size, followed by 3 to 15-μm AED and <3-μm AED cuts. At the time the experiment was started the EPA defined the inhalable particle cut-point at 15 μm AED. This was later changed to 10 μm. The SSI was evaluated for wind speed (1-10 mph) response (intake sampling bias) by Wedding (WE82) and McFarland (MC84) in wind tunnels and by RFP researchers during 50-100 mph winds in the field (RF3914, p4). Performance was satisfactory at low speeds, but at the high wind speeds some particles were apparently blown through the circular SSI inlet and out the opposite side without being sampled. The particle flux data from this scaffold represent the sum of all resuspension processes active in the 903 Field during each two month sample period. The long sample period is necessary to accumulate sufficient Pu for analysis. These data provide basic information to estimate possible population exposure and translocation of the Pu particles from the source area.

This research program extended from November 1982 thru August 1985, collecting bimonthly samples. This two month collection period was necessary to collect sufficient Pu in each size fraction for detection. The dust concentration data showed well defined trends with sampling height (RF4036, p21). However, the respirable dust particle concentration (8.0 $\mu g/m^3$) did not change with height, as expected for particles that are slowly sedimented. The concentration for the inhalable and coarse dust particles, ranging from 10 and 25 $\mu g/m^3$ respectively, dropped off with height due to sedimentation.

The Pu concentrations (RF4036, p21) for the <3-μm or, what are termed respirable particles, was 0.0088 fCi/m^3 of Pu-239 and for inhalable (3-15 μm) particles was 0.025 fCi/m^3 of Pu-239. These concentrations did not correlate with height, being only 3 and 10 times greater than background concentration respectively. One must keep in mind that the Pu mass for these samples represents about 1/10th part per billion of the total sample mass. Statistically the data have to be erratic at such extremely low concentrations. For a Pu concentration of 0.010 fCi/m^3, typical of the respirable Pu particle concentration at the scaffold, it would require the collection of just <u>one</u> 1-μm Pu particle per month by a hivol operating at 1.1 m^3/min.

The concentration of 0.067 fCi/m^3 of Pu-239 in the coarse (>15-μm) particle fraction was almost a magnitude higher than that in the respirable fraction (<3 μm). The coarse fraction exhibits a significant decrease in radioactivity with height by a factor of 3 from a height of 1 to 10 m.

No correlation was found with wind speed or direction for the Pu or dust concentration. This can be expected for the poor time resolution given by two month sample periods necessary to collect enough Pu for analysis (e.g., for the <3-μm particles).

It was obviously of interest to see how the Pu concentration changes beyond the 10 m scaffold. As a result, the ultravol sampler was operated in June 1981 at the cattle fence (Figure 2) 0.5 km due east of the scaffold. The Pu concentration in the inhalable (<10-μm) and coarse (>10-μm) ranges dropped off by a factor of 20, approaching background levels (UN81). Therefore, it did not seem worth while to continue this operation at such low levels. Evidently, most of the large Pu/soil particles that carry the bulk of the Pu radioactivity had settled out before reaching the cattle fence in the RFP buffer zone, far from any populated areas. This observation is directly supported by work of J. Hayden (HA75), who measured the size of individual Pu particles found on the soil surface from the 903 Field to Indiana Street, located 1.5 miles to the east (Figure 2). Beyond the cattle fence he considered RFP stack effluent to be the primary source of Pu particles because of the small size of these particles. The 1957 fire may have produced some small particles that settled out beyond the plant boundary. The total release was estimated at 26,000 μCi (EI80, p3-53).

Comparison of RFP Data with Previous Resuspension Studies

It is of interest to compare the above results to G. Sehmel's July 1973 Pu resuspension experiments at RFP (HA80c, p241). Although Sehmel's was a more elaborate Pu flux study than the study cited above, it only lasted for one month. Sehmel used three sampling scaffolds, one at the fence near the 903 Pad, one at the same location as the RFP 10 m scaffold and one near the cattle fence.

It is difficult to directly compare Sehmel's data to this study, because this study covered 34 months to determine statistically significant trends in the Pu concentration at three levels at one location. Sehmel's study probed the Pu particle plume at three locations with nearly 40 samplers set for specific wind speed ranges as well as continuous operation. However, to reiterate, Sehmel's research work was only of a month's duration.

There is also a problem comparing the particle fraction data. To achieve well-defined particle-size fractions, RFP took considerable precautions to coat the collection surfaces with adhesive. The object was to stop particles from bouncing through the cyclone and

cascade impactor stages onto the back-up filter (RF2866, p14; RF3115, p4). The SSI hivol inlet was also adhesive coated, long before this was an EPA requirement. Sehmel did not use adhesive on his collection surfaces. He showed that 60 to 99 percent of the Pu was in the respirable range and supposedly of RFP origin (HA80c, p262). We found that respirable Pu was mostly of fallout origin and it only represented 2 and 9 percent of the total Pu radioactivity, based on measurements at the scaffold and 903 Field respectively. Therefore, the Pu size trends are not comparable.

The drop off in Pu concentration as distance increased from the 903 Field security fence to the second scaffold varied among samplers by a factor of 10 to 1000 in Sehmel's tests (HA80c, p251). Comparable simultaneous data from our study were not available, due to access to only five hivol samplers. But taking data over the period 1980 to 1985, as RFP experiments moved eastward, the average Pu radioactivity at 1 m changed from 1.9 to 0.48, then to 0.13, and finally to 0.050 fCi/m^3 measured respectively in the 903 Field, just east of the field, the scaffold, and the cattle fence. This trend represents a 40-fold reduction over a distance of 1.5 km.

Sehmel did not report a definitive relationship between wind speed and Pu radioactivity (HA80c, p244). This is similar to RFP research experience in this area.

Isotopic Ratio Determination as an Indication of Long Distance Dispersion of RFP Pu

As a final test of whether any RFP Pu particles reach the general population, the mass isotopic ratio of Pu-240/Pu-239 was determined for a series of airborne particulate samples (RF4036, p22). RFP Pu production metal has a Pu-240/Pu-239 mass ratio of 0.051, while fallout has a ratio of 0.163. Airborne dust samples collected at the scaffold showed a ratio of 0.063 and nearby soil had a ratio of 0.054. This small difference was significant, indicating that the airborne dust carried some fallout Pu, as to be expected. It is now needed to obtain the isotopic ratios for air samples taken in various parts of the Denver region to identify the RFP contribution, if any, from 903 Field resuspension or production facility emissions.

For the latter program it is also necessary to take soil samples at the air sampler sites, since most background Pu (fallout Pu) now originates from resuspension of nearby soil particles (RF4036, p29). Stratospheric influx of Pu is very low at present. Therefore, the isotopic ratio of the soil should be known at the air sample sites to adjust for any RFP Pu, if any.

already in the soil, in addition to the fallout Pu from past nuclear weapons tests. Nearly 20 years ago isotopic ratios were determined by Krey (KR76, p213) for a limited number of soil samples in the greater Denver region to identify RFP Pu. This would also be a chance to determine if any changes occurred in these ratios. Krey's data show that 1 to 2 percent of RFP Pu deposited in the environment due to releases from the 903 Pad, plant stacks, and the 1957 and 1969 fires, extends beyond Indiana Street.

Resuspension Factors for Pu Release from the 903 Field

The resuspension factor (R_f) estimates the airborne contaminant concentration directly above a contaminated area and provides a means to estimate exposure or dose. R_f equals the airborne Pu concentration measured directly above a given area divided by the soil surface Pu concentration at that location. The necessary Pu data to calculate resuspension factors for the 903 Field are available. The soil surface Pu concentration is derived from the soil density and soil radioactivity per unit mass.

Another resuspension parameter is the resuspension rate (R_r), which allows off-site dose calculations. R_r is the fraction of the total radioactivity in the soil released per second. This provides a source term for meteorological calculations to determine downwind population exposure. R_f only provides dose or exposure for a person present on the contaminated area, which is somewhat academic for real life situations, because plant personnel only spend limited time on or near the 903 Field. The RFP Pu flux data make it possible to estimate R_r, but estimates of the Pu particle plume profile have to be made.

Before proceeding to estimate R_f and R_r, the limitations for applying these factors should be understood. Resuspension factors/rates ignore the physical parameters affecting resuspension, such as wind speed, vegetative coverage, soil moisture, precipitation and contaminant/host particle size. Also, a good knowledge of the Pu surface distribution is assumed, as well as airborne concentration over the whole area in question. As Sehmel (HA80c, p269) correctly points out, realistic prediction of the relationship between surface concentration and airborne concentration is fraught with uncertainties. Such data are very site specific and depend on how the contaminant found its way into the soil/vegetation and how long the contaminant has "weathered" into the soil. For example, resuspension for the first few weeks, after a tracer was sprayed onto cut grass, was orders of magnitude higher (RE79, p27) than the RFP data given below. The preference is to use actual Pu concentration data and then draw conclusions.

Sehmel (SE72) probably made the best estimates of maximum resuspension factors at RFP for a special situation in 1969, when Pu releases were high with no vegetation on the 903 Field during the remediation project that involved earth moving machinery. Samples were taken for time periods as short as six hours in the source area. The R_f values ranged from 10^{-9} to 10^{-5} m^{-1}. However, these factors are no longer applicable, unless similar areas of fresh soil are exposed.

An R_f range of 10^{-13} to 10^{-10} m^{-1} was calculated by us, limited to areas near the 903 Field sampler (RF4036, p44). The variability in soil Pu radioactivity (see Figures 3 and 4) and ground cover raises serious questions about generalizing from these values to the entire 903 Field. The same applies to the calculations for R_r, which was estimated at 2×10^{-12} sec^{-1} for the entire 903 Field area. This calculation required an estimate of the average air flow over the field and the resulting fetch for resuspended particles. This parameter was used to estimate the total emission of Pu from the field to be ~200 μCi per year.

The question of Pu transport to populated areas is better answered by downwind Pu concentration data provided earlier in this report. These long term measurements show that the Pu resuspended from the 903 Field does not contribute appreciably to off-site dose. The 903 Field influence beyond about 1.5 km could not be discerned. To further confirm this observation, future studies are suggested, involving additional air sampling along Indiana Street with improved air samplers that do a better job of efficiently collecting larger airborne particles (RF3650, p20). The Pu samples should be analyzed for the Pu-240/Pu-239 ratio as well as samples of nearby soil to identify their sources, such as fallout or Pu generated at RFP.

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